

**ULECAT**

CATALYTIC COMBUSTOR FOR ULTRA-LOW EMISSION DUAL FUEL GAS  
TURBINE

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# **ULECAT**

## **CATALYTIC COMBUSTOR FOR ULTRA-LOW EMISSION DUAL FUEL GAS TURBINE**

## ABSTRACT

The target of the project is to assess the feasibility of burning biomass-derived gases in a catalytic combustor which will replace the ordinary flame combustor, as a mean to overcome difficulties arising with the use of such fuels in gas turbines, in the range of 1 to 5 MWe. A dual fuel strategy is necessary for the practical use of gases generated from biofuels, because Diesel fuel will be used for start-up and back-up. It includes laboratory and pilot-scale investigations, as well as a preliminary technical and economic analysis of the system.

Hexaaluminates and noble metals catalysts as well as combinations of both are suitable materials for the dual-fuel combustor, while perovskites ought to be excluded for low durability reason. Ignition temperatures over base metal or noble metal catalysts are low enough so as to avoid a preheating system. The fuel-N conversion and the NO<sub>x</sub> formation have been studied extensively, and conversion of ammonia to molecular nitrogen ranging between 60 to 80 % have been achieved with low activity catalysts and in the 800 – 950°C temperature window. This conversion is probably due to a complex combination of SNCR and SCR processes, which were not clearly discriminated.

Pilot testing showed that low emissions of both unburned hydrocarbons and carbon monoxide could be achieved and for fuels with a low level of fuel bound nitrogen also low emissions of NO<sub>x</sub> could be achieved. But poor ammonia to molecular nitrogen conversion was achieved with the catalysts combination used.

The models developed to simulate the catalytic section of the combustor were used for the analysis of the pilot tests as well as for the preliminary design of the full-scale catalytic combustor.

Thermodynamic cycle calculations showed that a 35 % gas turbine efficiency can be reached with a machine running on biomass based gas, while the performance is a bit lower with Diesel. However, the global efficiency (including gas compression) is limited to 28 % with biomass based gas, and many configurations were investigated in order to improve this performance, among them the use of a recuperator seems to be the best solution.

The system analysis showed that the catalytic combustor might bring a real benefit regarding NO<sub>x</sub> emissions, with cost more than twenty to fifty times lower than SCR or scrubbing, as far as the fuel-N conversion is achieved. Nevertheless, if an additional NO<sub>x</sub> treatment is required, there is still a benefit if this treatment can be designed to have a limited performance (50 % for instance). Moreover, the catalytic combustion keeps an interest in stabilising the combustion of low-BTU fuels.

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## Objectives

Renewable fuels accounts for less than 2 % of the European Union primary energy requirements in the mid of the 90's, but advanced biomass power technologies as well as a new emphasis on short rotation crops and forest could change this pattern. A threefold higher biomass consumption is expected by 2005 and electricity production through Biomass Integrated Gasification Gas Turbine (BIGGT) will be probably the best short and medium term option for introducing this renewable resource into commercial energy channels, according to studies carried out with the support of the EU (ALTENER Program).

To limit transportation costs and negative effects, biomass ought to be processed locally. Therefore, BIGGT units will be of small- and mid-size, i.e. in the range of 1 to 50 MWe.

The use of biomass-derived fuels do not affect the net emissions of carbon dioxide in the atmosphere, and therefore prevents the greenhouse effect. In addition, sulphur dioxide emissions are limited due to the low sulphur content of biomass materials. The main concern related to pollutant emissions from a BIGGT unit are nitrogen oxides, carbon monoxide and unburned hydrocarbons. Regarding the aspect, the catalytic combustion appears as one of the most appropriate solutions to make a power generator totally free of pollutants.

The target of the project is to assess the feasibility of burning biomass-derived gases in a catalytic combustor which will replace the ordinary flame combustor, as a mean to overcome difficulties arising with the use of such fuels in gas turbines, in the range of 1 to 5 MWe. A dual fuel strategy is necessary for the practical use of gases generated from biofuels, because Diesel fuel will be used for start-up and back-up.

The catalytic combustor is one potential solution to overcome difficulties arising with the use of low-BTU fuels in a gas turbine. A more precise objective is to develop an ultra-low emission gas turbine able to run with both biomass-derived gases and liquid fuels. As typical values set as targets the following numbers have been used:

- Thermal efficiency >35 %
- UHC < 25 ppm at 15 % oxygen content

- CO < 50 ppm at 15 % oxygen content
- NO<sub>x</sub> < 25 ppm at 15 % oxygen content
- Fixed nitrogen conversion to NO<sub>x</sub> < 10 %

## **Technical description**

The program consisted in five main tasks:

- Task n°1 : Definition of the technical specifications
- Task n°2 : Development of new catalysts
- Task n°3 : Catalyst pilot-scale testing
- Task n°4 : Catalytic combustor modeling
- Task n°5 : Analysis of the system

### ***Task n°1 : Definition of the technical specifications***

This task consisted in a first analysis of the main aspects related to the use of biomass derived gases in a dual fuel gas turbine. It included the definition of the characteristics of the fuels (gases produced from gasification of wood and Diesel fuel) as well as a preliminary layout of the catalytic combustor and its operating conditions.

### ***Task n°2 : Development of new catalysts***

The aim of this task was to screen catalysts suited to comply the technical specifications. It included catalyst preparation, catalyst characterisation, lab-scale testing and kinetics studies. Active phases such as noble metals, hexaaluminates and perovskites have been investigated.

### ***Task n°3 : Catalyst pilot-scale testing***

The objective of this task was to validate the feasibility of catalytic combustion of biomass derived gases and Diesel fuel under realistic conditions, i.e. under actual space velocities, temperature, pressure and with real fuels. It included a validation of the synthetic mixtures used for lab-scale testing and pilot-scale testing with both fuels and with different catalysts prepared in task 2.

### ***Task n°4 : Catalytic combustor modeling***

Different models of the catalytic section of the combustor have been developed and validated by pilot-scale experiments. They provided a better understanding of the various physico-chemical phenomena governing the combustor behaviour and have been used for the final design of the combustor.

### ***Task n°5 : Analysis of the system***

This task consisted in an optimisation of the design of the combustor as well as a technical and economical analysis of the system. It included a comparison of the new developed technology over more conventional solutions.

## **Results and conclusions**

Task 1 led to a first layout of the catalytic combustor. Basically, this is an hybrid combustor with a first catalytic zone based on several catalytic segments and a second homogeneous zone for burnout of the gas. In addition, a pilot-combustor is placed after the catalytic zone, operating at low loads to start-up the gas turbine and at full load to increase the peak combustion temperature.

The results from Task 2 show that fuel-nitrogen conversion is a complex mechanism, including both heterogeneous and homogeneous reactions very dependent on reaction temperature, but also on catalyst formulation and fuel gas composition. The highest conversion of ammonia in the fuel to molecular nitrogen in the exhaust is reported at temperatures between 800 to 950 °C. Conversion of ammonia is believed to be initiated by oxidation to nitrogen oxide, followed by reduction of the nitrogen oxide by reactions with remaining ammonia and other fuel components, e.g. carbon monoxide, methane and higher hydrocarbons, to molecular nitrogen. Direct route from ammonia to molecular nitrogen is also possible, but probably less likely.

Since the conversion of ammonia is strongly dependent on the reaction temperature, it would be desirable to find operation conditions where the operating reaction temperature is limited to a narrow temperature window. The recuperative gas turbine could be a promising alternative with relatively high inlet temperatures and low outlet temperatures compared to a conventional gas turbine.

In task 3, a large number of different monolithic catalysts from KTH and IFP were evaluated but for comparison also commercial catalysts were utilised. The synthetic model gas mixture followed the same trend as the real gas but the differences in gas composition resulted in that the concentrations of unreacted CO and hydrocarbons did not totally overlap. Using the real gas in pilot scale tests it was shown that low emissions of both unburned hydrocarbons and carbon monoxide could be achieved and for fuels with a low level of fuel bound nitrogen also low emissions of NO<sub>x</sub> could be achieved.

The longest period used for the ageing tests was 50 hours of continuous operation on diesel fuel. After this period no decrease in catalytic activity could be determined. Neither for the deactivation tests using a fuel with high content of sulphur and aromatics could any decrease in activity be determined.

The testing using both fuels did not lead to any significant deactivation of the catalyst. After a number of rapid changes between the two fuels and a total of twenty hours of operation no significant difference in catalyst activity could be determined.

In task 4, different models of a single channel of the monolith segments have been developed by Politecnico di Milano and IFP. Comparison of model predictions with pilot scale experiments show a reasonable agreement. This was achieved by tuning of homogeneous combustion rates in the case of gasified biomass and by fitting of kinetic parameters for both heterogeneous and homogeneous rate expressions in the case of Diesel fuel. For this latter the effect of pressure (1-2.7 bars) was also

reproduced. Calculation of pressure drops in monoliths has been validated by comparison with cold flow experiments. Concentrated  $\Delta P$  coefficients and catalyst roughness emerged as the most critical parameters and have been fitted to match experimental data.

Constraint on pressure drops has been shown as the key parameter for sizing of the catalyst section. With the most conservative assumptions only 3-4 monolith segments (2.5 cm) can be used in the more critical case of gasified biomass. Adoption of advanced thin walled monoliths is beneficial.

With gasified biomass only partial combustion occurs in the catalyst section with significant CH<sub>4</sub> and CO outlet concentrations. Adoption of monoliths with high specific geometric area is beneficial but does not allow complete combustion in the catalyst section at partial load. Chemkin calculation performed at IFP showed that complete combustion can be achieved only with oversized post-combustion sections.

With Diesel fuel complete combustion is achieved in the catalyst section due to anticipated ignition of homogeneous combustion, but wall temperature exceeds 1000°C in the last segments. Ignition of homogeneous combustion is favoured by high cell densities.

In task 5, calculations made by Turbomeca and Volvo show that the required thermal efficiency of 35% for the gas turbine can be reached with biomass based gas for the Eurodyn turbine. The gas compression will however reduce the efficiency to about 28%. With a recuperative cycle a higher efficiency would be possible.

The economic analysis shows there is a considerable economic potential for catalytic gas turbine combustor in a biomass based IGCC compared with gas turbine with SCR (Selective Catalytic Reduction). For a mid size plant (approximately 60 MWe) the cost of NO<sub>x</sub> reduction with SCR is 40 times higher than the use of catalytic combustion. For a small size plant (approximately 3 MWe) the difference is even bigger. In this case it is however more correct to make the comparison with absorption of the ammonia (wet scrubbing) since probably no SCR is built for such a small plant. But also compared to the absorption technology, catalytic combustion would be an interesting alternative, provided that the NO<sub>x</sub> targets can be reached. Nevertheless, if the catalytic combustion does not fully meet the NO<sub>x</sub> limits as one can assume today, and if an additional NO<sub>x</sub> treatment is required, there is still a benefit if this treatment can be designed to have a limited performance (50% for instance). Apart from the fuel-N issue which requires additional investigations, the catalytic combustion keeps an interest in stabilising the combustion of low-BTU fuels.

## **Exploitation plans and anticipated benefits**

### **Context**

The greenhouse effect and more particularly the carbon dioxide emissions has never been a so topical question. Regarding power generation, a more intensive use of biomass as well as the achievement of higher energy efficiencies are two routes which could have a significant effect on reducing the CO<sub>2</sub> increase in the atmosphere.

Since the 80's, the United States have been making significant efforts in this direction and now, more than 10.000 MWe are biomass-based power generation. In Europe, a very limited number of power plants are fuelled by biomass, even if agricultural and forest residues are estimated above one billion tons per year in 1991. Therefore, it is crucial for the competitiveness of the European industry to make possible the use of biomass for electricity production. Costs must be mastered and

technical problems overcome regarding the whole route, e.g., feedstock preparation, biomass gasification, fuel-gas cleaning and power generation.

On the other hand, there is almost no doubt that gas turbine with catalytic combustor will be on commercial applications in a near future. The American company Catalytica, associated with several gas turbine manufacturers, is about to demonstrate the feasibility of the XONON combustion chamber, which aims at producing flue gases with NO<sub>x</sub> and UHC emissions below 3 and 10 ppm respectively. A 1.5 MWe demonstrator has been running successfully for more than 1000 hours. The marketing strategy of Catalytica is focused on small- and mid-size engines, with an emphasis on revamping. Provided that the catalytic combustion will certainly not introduce extra cost compared to most classical lean combustion strategies, this type of new engine will probably have a large market after 2000.

### **Technical and economic potential for exploitation**

The lab- and pilot-scale tests, as well as the analysis conducted by the gas turbine manufacturers show that there is no peculiar difficulties in applying the catalytic combustion to engines running on Diesel fuel and to meet the targeted values of pollutant emissions and efficiency defined at the beginning of the project.

The use of biomass-derived fuels in such a catalytic combustor is more complicated due to the gas composition, the presence of nitrogen-bound species and to the requirements of the catalytic combustion. Hence, the catalytic section operating temperature window is too narrow to avoid unburned products release at partial load. A solution is to adopt a cycle with a recuperator so as to reduce the temperature variation with respect to the load. The fuel-NO<sub>x</sub> issue is of greater importance, since no reliable strategy has been defined to maximise the fuel-N to molecular reaction, although a significant potential of 80% was identified at the lab-scale. Further investigations are needed to understand more clearly through which processes the 80% potential (or more) can be achieved, and then to study different combinations of fuel and air staging and catalysts arrangement that might provide the best compromise with constraints on the pressure drop, the wall temperature, the catalyst activity and the outlet temperature of the catalytic section.

Whatsoever, the catalytic combustion keeps an interest in stabilizing the combustion of very low-BTU gas and efforts have to be made on promoting it, even if light gas treatment for ammonia removal are necessary.

The preliminary investment evaluation of a 2.9 MWe cogeneration led to a 4000 Euros/kWe cost which is not acceptable in the current energy context, even if the feeds processed are available at very low prices. Significant efforts have to be made to reduce the cost of the most expensive equipments, gasifier, gas treatment, fuel handling, etc.

### **Actual applications targeted**

Since no real difficulties were met with Diesel fuel, a first objective is to pursue the development with this feed. Small and mid-size gas turbines are probably the most relevant target, before an enlargement to any size of gas turbine. To make sure to get a significant market, both new machines and revamping operations must be considered. Regarding the position of competitors, mainly in the USA and in Japan, a demonstrator ought to be shortly designed, erected and run so as to address issues such as durability, control strategy, etc.

As the ULECAT project was not completely successful concerning the use of biomass-derived fuels, additional basic research has to be carried out so as to enlarge the operating temperature window and thus to make the control of the combustor less

critical at any load, especially with respect to unburned products emissions. That means development of catalyst formulations which might withstand temperatures higher than the current limit of 950 – 1000 °C. A gain of 50 to 100 °C would have a significant impact on the reliability of the catalytic combustor.

The other important topic which has to be addressed on a fundamental basis is the fuel-NO<sub>x</sub> reduction. Again, an additional program has to be launch whose aim would be to get information about how the conversion of fuel-N to molecular reaction proceeds and then to validate on pilot units strategies maximizing this reaction.

These two topics are of great importance, regarding the growing interest of the final users for alternative cheap fuels or waste gaseous fuels.

It is important to mention that any new gas turbine in the future ought to be equipped with a recuperator in order to achieve acceptable efficiency, especially regarding other options such as reciprocating engines. Even if some development has already be made on this equipment, it is not yet commercially available, and additional work has to be carried out, essentially at the demonstration level.

Work is also underway to elaborate new schemes dedicated to small cogeneration units, enabling to reduce the investment cost below 2000 Euros/kWe, with a special emphasis on thermolysis which produces a mid-BTU gas easier to handle in gas turbines and giving higher global efficiency.